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Study of Infrared Nonlinear Processes in Semiconductors

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This program includes:

1.) Demonstration that phase transitions and electrical instabilities enhance optical nonlinearities in semiconductors.
2.) Discovery of exceedingly large third order optical nonlinearities [$\chi^{(3)} > 10^{-3}$ esu] with picosecond response time in zero gap materials (HgTe, HgCdTe, HgMnTe).
3.) First observation of impurity-induced optical nonlinearity in semiconductors.
4.) Prediction and observation of negative absolute carrier mobilities in quantum wells.

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I. Research Objectives and Achievements

This is the final technical report for AFOSR Grant 85-0269, entitled "Study of Infrared Nonlinear Processes in Semiconductors." The primary aim of this program was to discover materials and/or structures with large, fast nonlinear optic susceptibilities. Such elements are required in optical signal processing systems, and for protection of imaging devices. The program was primarily experimental, with supporting theoretical work. Tests of nonlinear crystals provided important information concerning carrier kinetics in semiconductors, through the difference frequency variation of $\chi^{(3)}$. We have used this technique to measure carrier relaxation times — in the picosecond range — in n-Si:P, HgTe, HgCdTe, HgMnTe, and HgCdSe:Fe.

The major achievements of the program include:

1.) Demonstration that $\chi^{(3)}$ is enhanced at the metal-insulator transition in n-Si:P.

2.) Discovery of exceedingly large ($\chi^{(3)} \approx 10^{-3}$ esu) optical nonlinearities with picosecond response times in zero-gap materials (HgTe, HgMnTe, HgCdTe).

3.) First observation of impurity-induced optical nonlinearities in semiconductors.

4.) Prediction and observation of negative absolute carrier mobilities in quantum wells.

Specific projects are discussed in more detail below.

II. Free Carrier, Spin-Induced Faraday Rotation in HgCdTe and HgMnTe

Electrons in narrow-gap semiconductors have large g-values and large spin-photon interactions. When their spins are aligned by a magnetic field $\left(\langle\sigma_z\rangle \approx 1\right)$ this interaction causes Faraday rotation, whose Verdet constant has a sharp resonance as the optical energy approaches the band gap. Thus, the process can be used as a band pass filter to select near-gap radiation.

We have studied spin-induced Faraday rotation of CO_2 laser radiation in n-HgCdTe and n-HgMnTe with E_G = 110-130 meV. At T = 2 K, the Verdet constant at resonance in an n-HgCdTe crystal containing 10^{15} electrons/cc is 0.19 degree/cm-gauss. This value is ten times larger than that expected from interband Faraday rotation, and two orders of magnitude larger than the free carrier plasma effect. The Verdet constant of HgCdTe decreases with laser intensity. In a lightly doped n-HgMnTe crystal, the Verdet constant is 1.8 degree/cm-gauss. This value exceeds that predicted theoretically by a factor of five.

In both HgCdTe and HgMnGe, the Faraday rotation rapidly decreases as the light is tuned away from the band edge. For example, in one case the rotation at 10.6 μ was only 25% that at 9.5 μ.

The resonant Faraday effect may find application in laser isolators or tunable IR filters. High quality HgCdTe crystals were provided by Dr. D. Nelson of Honeywell, Lexington. This work was reported at the 1986 MCT Workshop in Dallas.

III. Nonlinear Optics at the Metal-Insulator Transition

Four wave mixing experiments have demonstrated that heavily-doped n-Si:P samples ($n \approx 10^{18}$/cc) have large nonlinear optic susceptibilities ($\chi^{(3)} \approx 10^{-7}$esu) at 10.6 μ. This result is striking since lightly doped n-Si is one of the most linear semiconductors, with $\chi^{(3)} \lesssim 10^{-11}$ esu. Even in n-Si crystals with $n = 10^{17}$/cc, the nonlinear susceptibility, per carrier, is 10 times smaller than in those with $n = 10^{18}$ cc. These results suggest that the large $\chi^{(3)}$ of n-Si:P is a consequence of the metal-insulator (M-I) transition, which occurs at $n_c = 3.7\times10^{18}$/cc in n-Si:P.

To test this idea, we have developed a model of the nonlinearity; it postulates that the effect is caused by carrier temperature modulation produced by a combination of the two laser fields. Near the M-I transition, the temperature modulation promotes carriers from localized to delocalized states, thereby modulating the dielectric properties of the system. The model predicts the observed $\chi^{(3)}$; it also leads to an important formula for the contribution of each electronic state to the dielectric function. The expression varies, by more than a factor of 20, between localized and delocalized states.

These studies have elucidated the mechanism of impurity-induced optical nonlinearity in semiconductors. They also suggest that optical nonlinearities can be enhanced by phase transitions or electrical instabilities. Work along these lines, under item VI, was stimulated by the n-Si:P experiments.

IV. Optical Nonlinearity of Zero Gap Semiconductors

In 1986 we demonstrated a room temperature $\chi^{(3)} \simeq 10^{-4}$ esu, with 5 picosecond response time, in a HgTe epilayer. At the time, that was the largest known nonlinear optic susceptibility with picosecond speed. Since then, the susceptibility has been further increased, to 1.5×10^{-3} esu, in $Hg_{0.84}Cd_{0.16}Te$ which has a very small ($\simeq$ 7 meV) open band gap at 80 K. Here, again, the response time (as determined from the frequency dispersion of $\chi^{(3)}$) is 5 psec. The measurements were performed with finite-difference-frequency and degenerate four wave mixing experiments.

Electron-hole generation, via transitions between the Γ_6 and Γ_8 bands, is responsible for the nonlinearity of these small gap semiconductors. The effect is magnified by the large interband absorption coefficient ($\alpha \simeq 3000$ cm^{-1}) and small conduction band mass. Population modulation is caused by modulation of the electron-hole plasma temperature, relative to that of the lattice, with electron and hole quasi-Fermi levels <u>remaining equal to one another</u>. This situation is required by the very short ($\simeq 10^{-13}$ sec) electron-hole recombination times in zero-gap materials. As a consequence, the mechanism of their optical nonlinearity is substantially different from that of open gap semiconductors, where recombination times are long. Though conventional band filling gives rise to huge $\chi^{(3)}$'s (in materials such as InSb), they are slow and saturate easily. By contrast, the zero-gap nonlinearities are fast (5 psec), and only begin to saturate at laser intensities of 100 kW/cm^2 (1000 times that of convential band filling).

MBE-grown epilayers for this work were provided by Professor J.F. Schetzina of N.C. State University. We anticipate a continuing collaboration with his group.

V. Nonlinearities due to Resonant Impurity Levels

Energy-dependent scattering gives rise to electrical, and possibly optical, nonlinearities in semiconductors. The latter, however, require "abrupt" carrier scatterings; optical nonlinearity is not produced by relatively "soft," ionized impurity scatterings. Until recently, no simple semiconductor system meeting these requirements was known. The situation changed when Mycielski developed (HgCdFe)Se. In this crystal the Fe^{2+} level can lie within the conduction band, acting as both a donor and sharply resonant scatterer. This unique situation is ideally adapted to nonlinear optics. In samples with the resonance in the band, we find $\chi^{(3)}$'s that exceed those with it in the gap by a factor of 20. Measured susceptibilities are in agreement with those predicted by a theory based on the Anderson model. The measurement were performed with samples kindly provided by Professor Mycielski; these studies will continue with crystals now being grown at MIT. With appropriate doping and band gap we anticipate $\chi^{(3)}$'s exceeding 10^{-5} esu.

VI. Instability-Enhanced Optical Nonlinearity

Our observation of a sizable optical nonlinearity at the metal-insulator transition in n-Si:P (see Item III) suggests that phase transitions and electrical instabilities might generally be used to increase optical nonlinearity. Semiconductors exhibit a variety of dc transport nonlinearities that sometimes give rise to instabilities and domain formation. We have demonstrated, theoretically, that i-v characteristics of the S-shaped type can be used to enhance optical nonlinearity. In particular, the calculations imply that, when biased to the instability threshold, the low frequency ($\Delta\omega$=0) nonlinear susceptibility of a semiconductor diverges; the divergence is similar to those observed in light scattering experiments at second order phase transitions. An experiment to test this idea in cold, n-Ge gave a surprising and fascinating result. The sample was reproducibly switched — by the CO_2 laser optical pulses — back and forth between two bistable states of the electrically driven Ge crystal. Such an effect could serve as the basis for an optically-addressed spatial light modulator. The n-Ge experiments are continuing, with a view to understanding the mechanism for optical switching. In addition, we are testing an integrated GaAs structure (memo attached) that is expected to exhibit electrical bistability, and be optically addressable.

VII. Negative Carrier Mobilities in GaAs Quantum Wells

In collaborative work with the AT&T Bell Laboratories group, we have demonstrated that minority carriers optically injected into GaAs quantum wells can have negative absolute mobilities. For example, in p-type QW's, electrons injected into the structure flow toward the negatively biased electrode at low temperatures. The effect is caused by friction between the drifting electron and hole gases. Measurements of the minority carrier mobility can be used to infer the minority-majority momentum relaxation times. Experimentally, there is a large asymmetry between the cases of minority electrons in p-type quantum wells and minority holes in n-type quantum wells. In the former, the electron-hole momentum relaxation time is 60 fsec; in the latter it is 4 psec.

VIII. Publications

1.) "Hole Induced Four Wave Mixing and Intervalence Band Relaxation Times in p-GaAs and p-Ge," S.Y. Yuen, P.A. Wolff, L.R. Ram-Mohan, and R.A. Logan, Solid State Commun. 56, 489 (1985).

2.) "Nonlinear Optics near the Metal-Insulator Transition," P.A. Wolff, S.Y. Yuen, and G.A. Thomas, Solid State Commun. 60, 645 (1986).

3.) "Free-Carrier Spin-Induced Faraday Rotation in HgCdTe and HgMnTe," S.Y. Yuen, P.A. Wolff, P. Becla, and D. Nelson, J. Vac. Sci. Tech. A5, 3040 (1987).

4.) "Optical Nonlinearity in Mercury Telluride," P.A. Wolff, S.Y. Yuen, K.A. Harris, J.W. Cook, Jr., and J.F. Schetzina, Appl. Phys. Lett. 50, 1858 (1987).

5.) "Optical Nonlinearity in $Hg_{0.84}Cd_{0.16}Te$ and $Hg_{0.97}Mn_{0.03}Te$," S.Y. Yuen, P.A. Wolff, K.A. Harris, J.W. Cook, Jr., and J.F. Schetzina, J. Vac. Sci. Tech. (in press).

6.) "Free-Carrier Induced Optical Nonlinearities in Semiconductors," P.A. Wolff, S.Y. Yuen, K.A. Harris, J.W. Cook, Jr., and J.F. Schetzina, SPIE Conference Proceedings (in press).

7.) "Optical Nonlinearity due to Resonant Impurity Scattering of Electrons in HgCdSe:Fe," P.A. Wolff, S.Y. Yuen, R.R. Galazka, and A. Mycielski, J. Vac. Sci. Tech. (in press).

8.) "Negative Absolute Mobility of Minority Electrons in GaAs Quantum Wells," Ralph A. Höpfel, Jagdeep Shah, Peter A. Wolff, and Arthur C. Gossard, Phys. Rev. Lett. 56, 2736 (1986).

9.) "Negative Absolute Mobility of Holes in n-doped GaAs Quantum Wells," R.A. Höpfel, J. Shah, P.A. Wolff, and A.C. Gossard, Appl. Phys. Lett. 49, 572 (1986).

10.) "Electron-Hole Scattering in GaAs Quantum Wells," R.A. Höpfel, J. Shah, P.A. Wolff, and A.C. Gossard, Phys. Rev. B (in press).

IX. Theses

"Magnetic Polarons in Semimagnetic Semiconductors — a Time-Resolved Photoluminescence Study of Exciton-Polaron Complexes in $Cd_{1-x}Mn_xSe$ and $Cd_{1-x}Mn_xTe$," John J. Zayhowski, Ph.D. Thesis in Electrical Engineering and Computer Science, MIT, February 1986.

"Light Scattering and the Bound Magnetic Polaron," Eric David Isaacs, Ph.D. Thesis in Physics, MIT, December 1987.

X. Personnel

Prof. P.A. Wolff, Principal Investigator
Dr. S.Y. Yuen, Co-Principal Investigator
Prof. L.R. Ram-Mohan, Consultant
E. Isaacs, Physics Graduate Student
C. McIntyre, Physics Graduate Student
J. Stark, Physics Graduate Student
D. Walrod, Physics Graduate Student
X. Wang, Physics Graduate Student
S. Wong, Physics Graduate Student
J. Zayhowski, Electrical Engineering and Computer Science Graduate Student

XI. Interactions

We have a continuing interaction with AT&T Bell Laboratories through Professor Wolff's consulting contract. Dr. G.A. Thomas, of Bell, provided samples and was a co-author of our paper on optical nonlinearities in n-Si. One of our students, J. Stark, spent the summer of 1986 at Bell Laboratories, Holmdel, fabricating structures for high field transport measurements in (HgCd)Se:Fe. His research on nonlinear transport and pico-second optics is continuing under the joint supervision of Professor Wolff and Dr. D. Chemla (Bell Labs, Holmdel).

We collaborate with the Honeywell Electro-Optics Group (Lexington, MA) in experiments requiring HgCdTe crystals. Drs. M. Reine and D. Nelson have been very helpful in providing high quality samples.

The zero gap (HgTe) work is collaborative with Prof. J. Schetzina's group (N.C. State University). He is now providing us (HgCd)Te epilayers and HgTe/CdTe superlattices to optimize the zero gap nonlinearities.

At MIT we interact regularly with Dr. P. Becla and Professor A. Witt regarding growth and characterization of semiconductor crystals. We also discuss nonlinear optical processes and devices with Professors E. Ippen and H. Haus.

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MEMORANDUM

To: R. Aggarwal, P. Becla, C. Fonstad, B. Goldberg, D. Heiman, E. Isaacs, C. McIntyre, L.R. Ram-Mohan, J. Stark, S. Wong, S. Yuen
From: P.A. Wolff -PAW
Subj: Bistable Electronic Device
Date: November 25, 1986

This memo proposes a simple, bistable electronic device that can be scaled to small dimensions (<1μ), will be easy to fabricate, and has the potential to be switched both electrically and optically. The device achieves bistability via the Gunn effect. As is well known, in bulk n-GaAs crystals the negative differential conductivity (NDC) associated with the Gunn effect leads to domain formation and oscillation. We suggest that in a suitably doped n-GaAs microstructure, NDC will produce two different stable field configurations whose sense could be determined electrically. Voltage pulses, and possibly optical pulses, could be used to switch the device between its two stable states.

Consider an n-type GaAs epilayer doped in the pattern illustrated below:

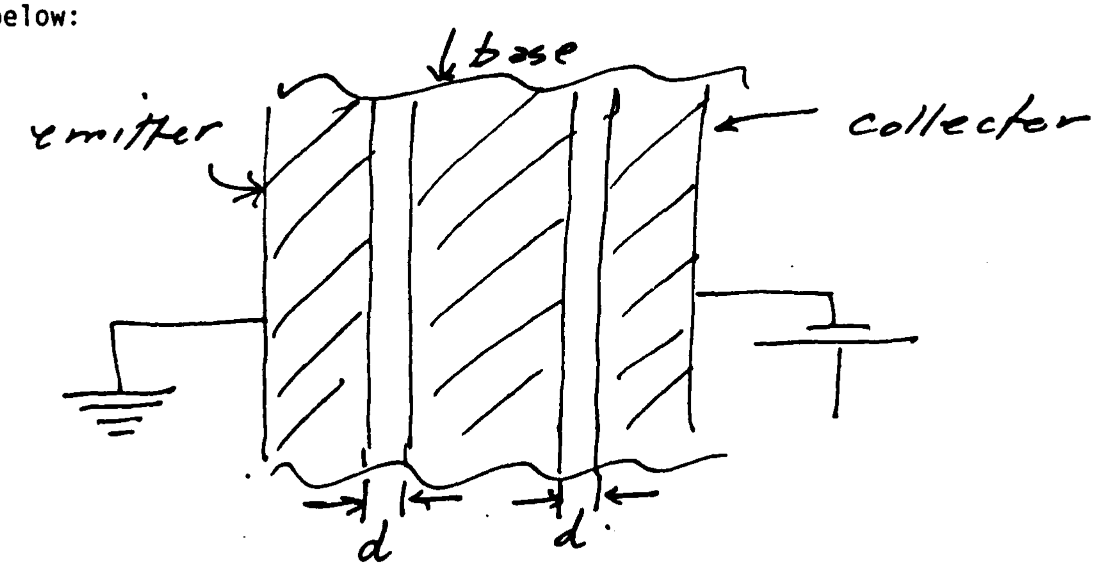
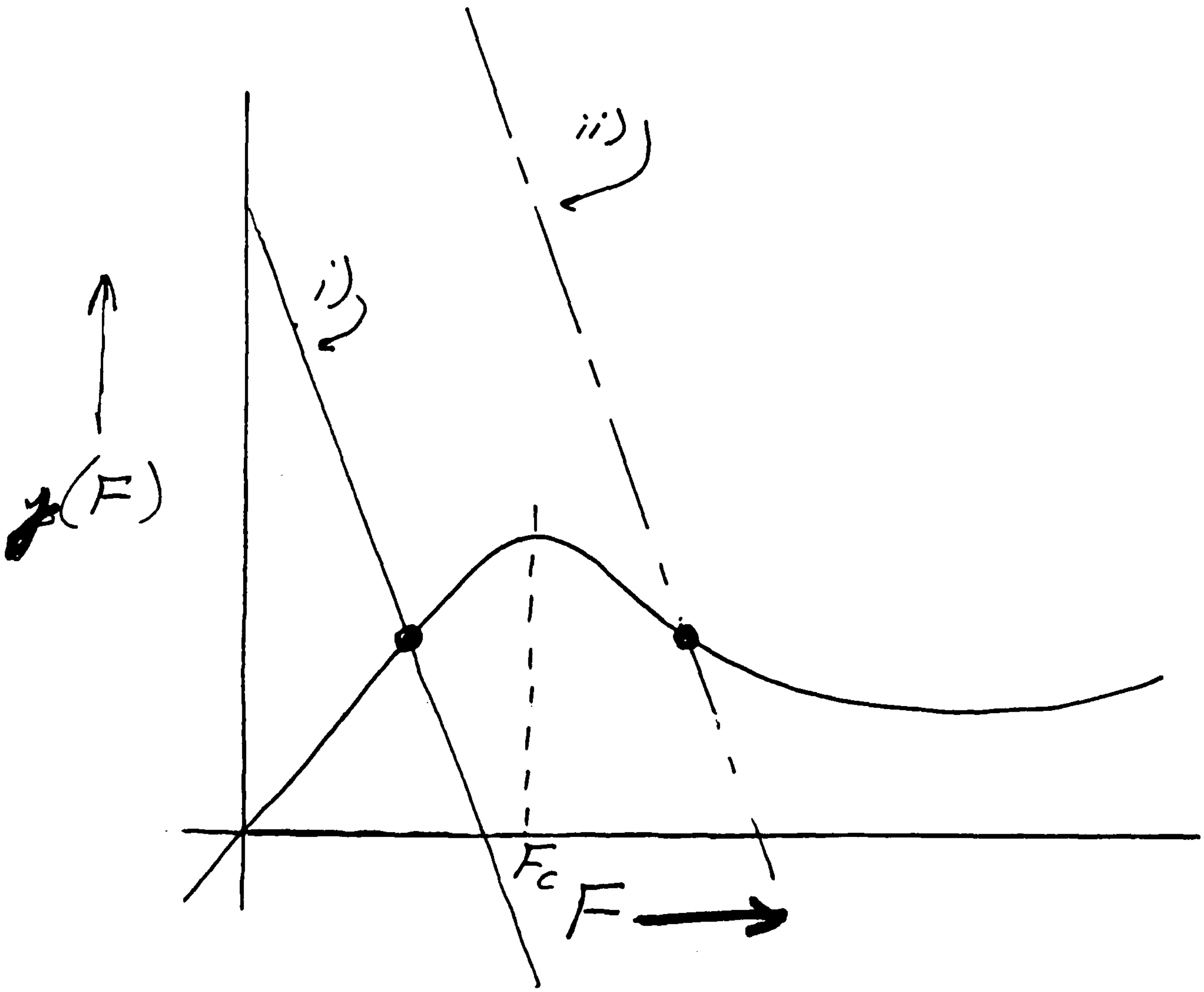


Fig. 1

Cross hatched regions are heavily doped ($n \simeq 10^{17}/cc$); unshaded regions are lightly doped ($n \simeq 10^{14}/cc$). The thickness (d) of the lightly doped regions is chosen sufficiently small that Gunn domains <u>cannot</u> develop within them. Smith[1] gives the condition nd > 2 x 10" cm^{-2} for domain formation; thus, with $n = 10^{14}$ cm^{-3} and d = 1μ, domain formation should be suppressed. This condition will allow us to assume that the electric field is uniform in each of the lightly doped, active regions. However, we will not require that the field (F_1) in region 1 be equal to that (F_2) in region 2.

We now investigate the electrical response of the device when the collector is biased with respect to the emitter, and the base floats. Since the lightly doped layers have high resistivity, most of the voltage drop falls across them i.e. $V \approx (F_1 + F_2)d$. Initially we assume that $F_1 = F_2$, and consider the stability of such a field division. The current is then determined by the intersection of the current-field characteristic of the lightly doped regions with the load line[2]. The figure below illustrates two cases; i) an applied voltage below that required to produce NDC and ii) a voltage in the NDC regime.



In case i), the load line intersects the j-F characteristic at a point where $(dj/dF) > 0$. We will show below that the equal division of fields ($F_1 = F_2 \approx V/2d$) is then stable, and the base floats at potential $V_B = V/2$. On the other hand, at higher voltages ($V/2d > F_c$) the intersection occurs in a region of NDC, and the $F_1 = F_2$ field distribution is unstable. These statements can be intuitively understood by considering the effect of a small fluctuation in the base potential, $V_B \to V/2 + \delta V$. The fluctuation increases the field in region 1 ($F_1 \to V/2d + \delta V/d$) and decreases that in region 2 ($F_2 \to V/2d - \delta V/d$). The resulting change in the currents $[j_1 \to j + (dj/dF)(\delta V/d),\ j_2 \to j - (dj/dF)(\delta V/d)]$ decreases the fluctuation if $(dj/dF) > 0$, but amplifies it in the NDC region where $(dj/dF) < 0$.

Ridley's[2] analysis of the stability problem can easily be extended to the present case. By combining Poisson's equation;

$$\nabla \cdot F = (4\pi/\varepsilon)\rho \tag{1}$$

with the continuity equation,

$$d\rho/dt + \nabla \cdot j = 0, \tag{2}$$

he finds

$$\nabla \cdot \dot{F} = -(4\pi/\varepsilon)\nabla \cdot j. \tag{3}$$

In the one dimensional geometry we are considering, Eq. (3) can be integrated from region 1 to region 2 to give the result:

$$(\dot{F}_2 - \dot{F}_1) = -(4\pi/\varepsilon)\,[j(F_2) - j(F_1)]. \tag{4}$$

Finally, if $F_2 = (F + \delta F) \equiv (V/2d + \delta F)$ and $F_2 = (F - \delta F)$, the field fluctuation (δF) satisfies the equation:

$$\delta\dot{F} = -(4\pi/\varepsilon)(dj/dF)\,\delta F. \tag{5}$$

Field fluctuations decay when $(^{dj}/_{dF}) > 0$, but grow when the structure is biased into the NDC regime $(^{dj}/_{dF}) < 0$. In the latter case, it will come to steady state with <u>unequal</u> fields in the two active layers. For a symmetric device, such as that illustrated in Figure 1, there are then two equivalent steady state field distributions.

<u>Field Determination in the NDC Case.</u>

We now relax the condition $F_1 = F_2$ to determine the fields and current in the NDC case. In steady state, we require

$$j(F_1) = j(F_2) = j \tag{6}$$

and

$$V = (F_1 + F_2)d + (jR) \tag{7}$$

where R is the resistance, per square centimeter, of the emitter plus base plus collector regions. To analyze these equations it is convenient to introduce new variables:

$$X \equiv 1/2(F_1 + F_2) \tag{8}$$

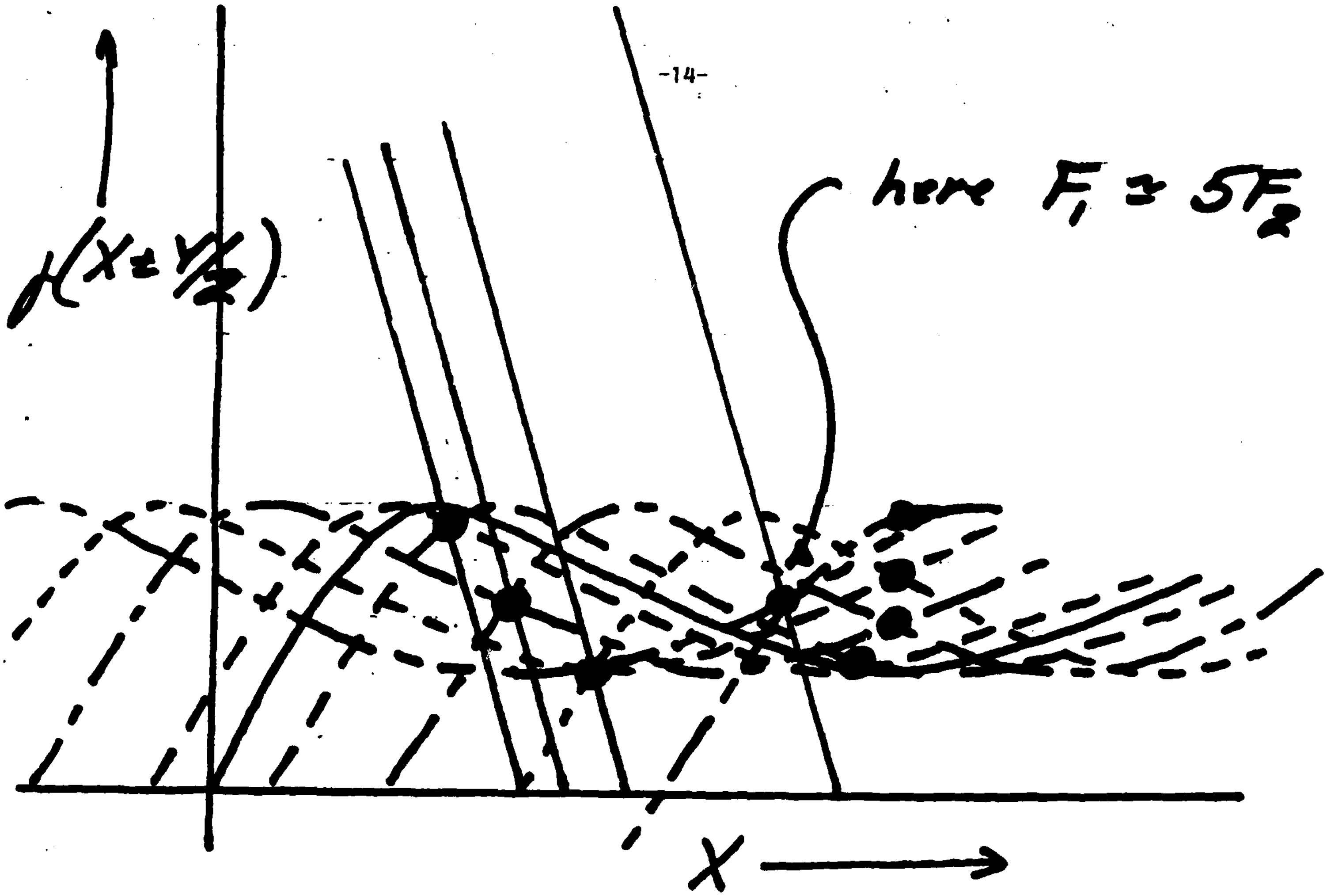
$$Y \equiv (F_1 - F_2) \tag{9}$$

Eqs. (6) and (7) then take the form:

$$j(X+^{Y}/_{2}) = j(X-^{Y}/_{2}) = R^{-1}(V-2Xd). \tag{10}$$

The last factor in this equation is the load line; it has a large (negative) slope because R is small.

To determine X and V, for a given value of Y, we plot the functions $j(X+^{Y}/_{2})$ and $j(X-^{Y}/_{2})$ versus X. The following figure illustrates such plots for three values of Y.



Note that for each value of Y (unequal to zero) there are two (or zero) intersections of the curves $j(X+{}^{Y}/_{2})$ and $j(X-{}^{Y}/_{2})$. The voltages required to achieve these solutions are determined by passing a load line, of appropriate slope, through the intersections. We note that at the intersection of largest Y, $F_1 \approx 5\,F_2$. The corresponding solution with $Y \to -Y$ has $F_2 \approx 5\,F_1$. These are the two bistable states of the device.

Their stability can be confirmed from Eq. (4) by making the replacements $F_1 \to (F_1-\delta F)$, $F_2 \to (F_2+\delta F)$. The equation governing the field fluctuation then takes the form:

$$2\delta F = -({}^{4\pi}/_{\epsilon})\ [{}^{dj}/_{dF}|_{F2} + {}^{dj}/_{dF}|_{F1}]\ \delta F. \qquad (11)$$

It is easy to see that the lower voltage intersections give <u>positive</u> values for the sum $[{}^{dj}/_{dF}|_{F2} + {}^{dj}/_{dF}|_{F1}]$. Thus, the corresponding solutions truly are <u>bistable</u>.

Switching

To electrically switch the device between its two stable states, voltages are applied to the base region. For example, if the device is in the state with high field between the base and collector, it can be reversed by bringing the base to the collector potential, V. Alternatively, if the high field domain is between the emitter and base, the device is reversed by grounding the base.

Optical switching might be achieved by taking advantage of the Franz-Keldysh effect, which effectively reduces the band gap in the high field domain. By pumping the structure with a burst of slightly-below-gap light, it should then be possible to selectively excite electron-hole pairs in the high field region. This charge would short out the high field, thereby forcing the voltage drop to the other active region of the device, and reversing it.

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